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"STUDIES OF THE DELAYED NEUTRONS"

by

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STUDIES OF THE DELAYED NEUTRONS

I. The Decay Curve and the Intensity of the Delayed Neutrons

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ABSTRACT

The delayed neutrons resulting from slow neutron fission of uranium have been found to decay as a combination of exponentials with half-lives of 0.4, 1.8, 4.4, 23 and 56 seconds and respective initial relative intensities when activated to saturation of 0.4, 0.5, 1.1, 1.0 and 0.14. Under equilibrium conditions the delayed neutrons are $1.0 \pm 0.2\%$ as abundant as the instantaneous fission neutrons.

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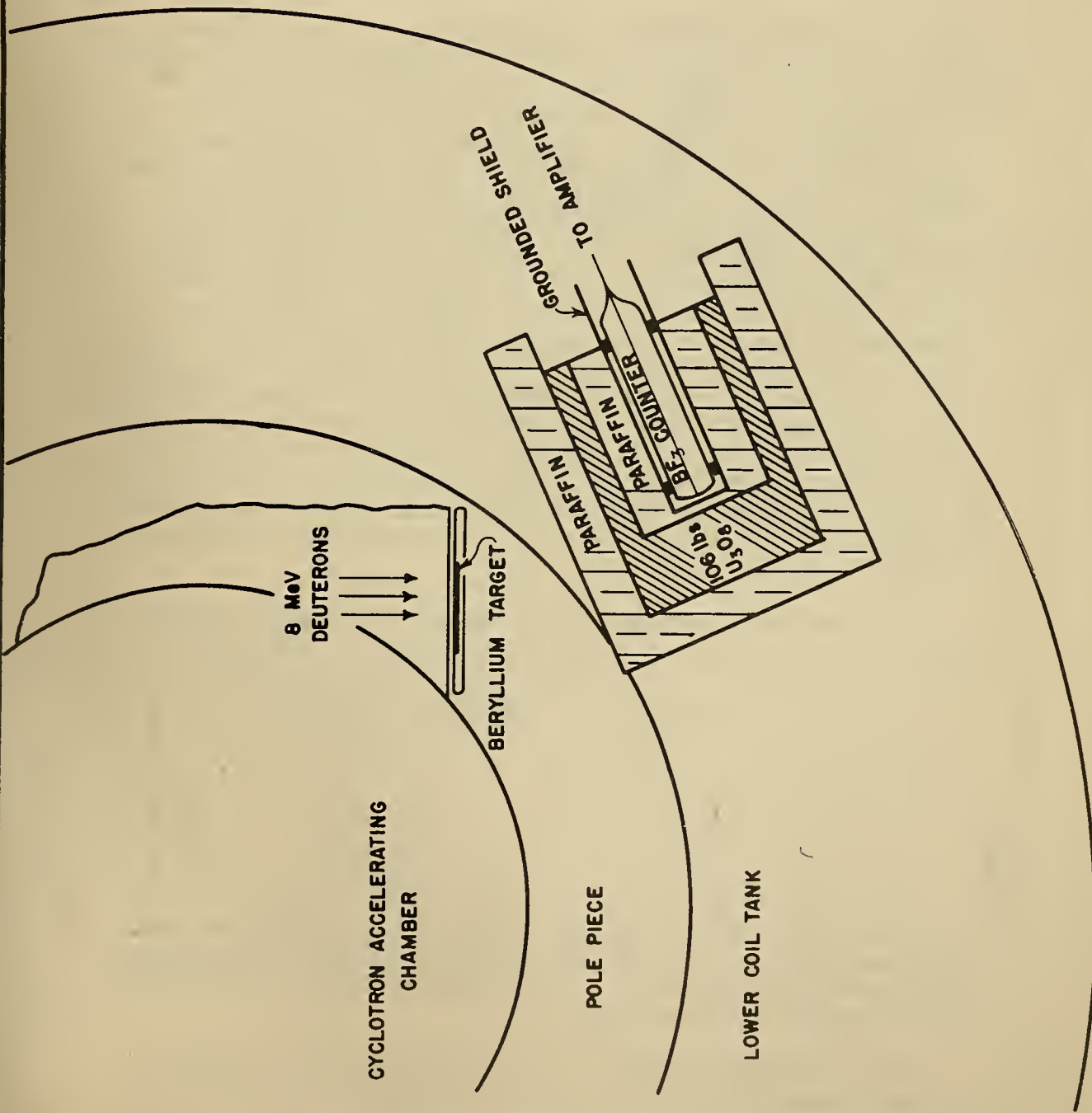
More than a year before the first nuclear chain reaction was initiated on December 2, 1942, the importance of the delayed neutrons in controlling the reaction was foreseen. In a letter to S. K. Allison, E. Fermi pointed out the necessity of knowing how many of the fission neutrons were delayed, and suggested how this could be measured experimentally. The work was undertaken, using the University of Chicago cyclotron, and it culminated in a Metallurgical Laboratory report by Snell, Nedzel and Ibser, the results of which have since been quoted in the Smyth report¹. As a part of the intensity measurement, a study of the decay curve for the delayed neutrons was required. While this was accomplished in a manner adequate at the time, subsequent work in the same laboratory showed that the analysis of the short periods had been incomplete. It is the purpose of this paper to state the revised analysis of the delayed neutron decay curve, and to describe the intensity measurement more fully than was done by Smyth.

The Decay of the Delayed Neutrons

The earlier work on this subject was that of Roberts, Mayer, Hafstad and Wang² (decay period 12.5 ± 3 sec.), of Booth, Dunning and Slack³ (10-15 seconds and 45 seconds), of Gibbs and Thomson⁴ (no strong delayed neutron periods between 0.001 and 0.1 sec), and of Boström, Koch and Lauritsen⁵ (12.3 and 0.1 - 0.3 second activities). Snell, Nedzel and Ibser used 106 lb of U_3O_8 arranged in a hollow shell which was surrounded with paraffin and placed about a foot from the beryllium target of the cyclotron (Fig. 1). A boron trifluoride proportional counter was placed inside the shell, and a thickness of $2\frac{1}{2}$ inches of paraffin filled the space between the counter and the U_3O_8 . The maximum cyclotron beam on the target was 20 μ A of 7.5 Mev deuterons. Readings were taken by photographing simultaneously the mechanical recorder actuated by the scaler, the scaler interpolation lights, a stopwatch which read to 1/100 second, and an electrical timer. The clearest result was the appearance of two well-defined activities with half-lives of 24 sec and 57 sec respectively. In relative saturation intensity, the 24 sec activity was about eight times as strong as the 57 sec activity. The earlier part of the decay curve could be accounted for by activities with half-lives of 7 sec and 2.5 sec., but the analysis was not definite enough to enable one to have confidence in these as real activities.

In later work upon the shorter periods, activations of about 1 second were given, and the readings were taken without automatic recording. Instead, one man would call time intervals, while another would read the counts and a third would record them. With a little practice this method of operation seemed to work well. It had the advantage that the data could be taken and analysed more rapidly than was possible with photographic film. By standardizing irradiation and counting times, repeated runs

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could be made which could be averaged simply. The main result of this work was the emergence of a fairly intense, well-defined activity with a half-life of 4.5 seconds. The shorter activity seemed to have a half-life of about 1.5 seconds, and the values of the half-lives of the previously-discovered longer periods were modified to 23 seconds and 56 seconds respectively.

In a third phase of the experimentation, rapidly circulating uranyl nitrate solution was used. The stream passed from a reservoir through a two-stage turbine pump driven above its rated speed by a 5 H. P. motor, through an irradiation cell near the target of the cyclotron, and thence past two BF_3 counters in series and into a sump tank. The counters were surrounded with paraffin and massively shielded against direct cyclotron neutrons. The flow time between the two counters could be varied by altering the volume of tubing between them, or by by-passing some of the solution around the second counter. The first counter was a monitor; to a first approximation the flow time to it from the irradiation cell was constant. With this apparatus, timing was reduced to measurements of volume and rates of flow. Irradiation lasted 0.19 sec and the first point on the decay curve was at a time 0.32 sec after the liquid left the irradiation cell. At this and subsequent chosen points one could take as many counts as one desired. The results confirmed the presence of the 4.5 sec period but gave for the half-life the value 4.4 sec; they indicated also an activity with a half-life of 1.8 sec. The first two points on the decay curve showed that a shorter period was also present; it could not be evaluated accurately, but had a half-life of about 0.4 sec and a moderate saturation intensity.

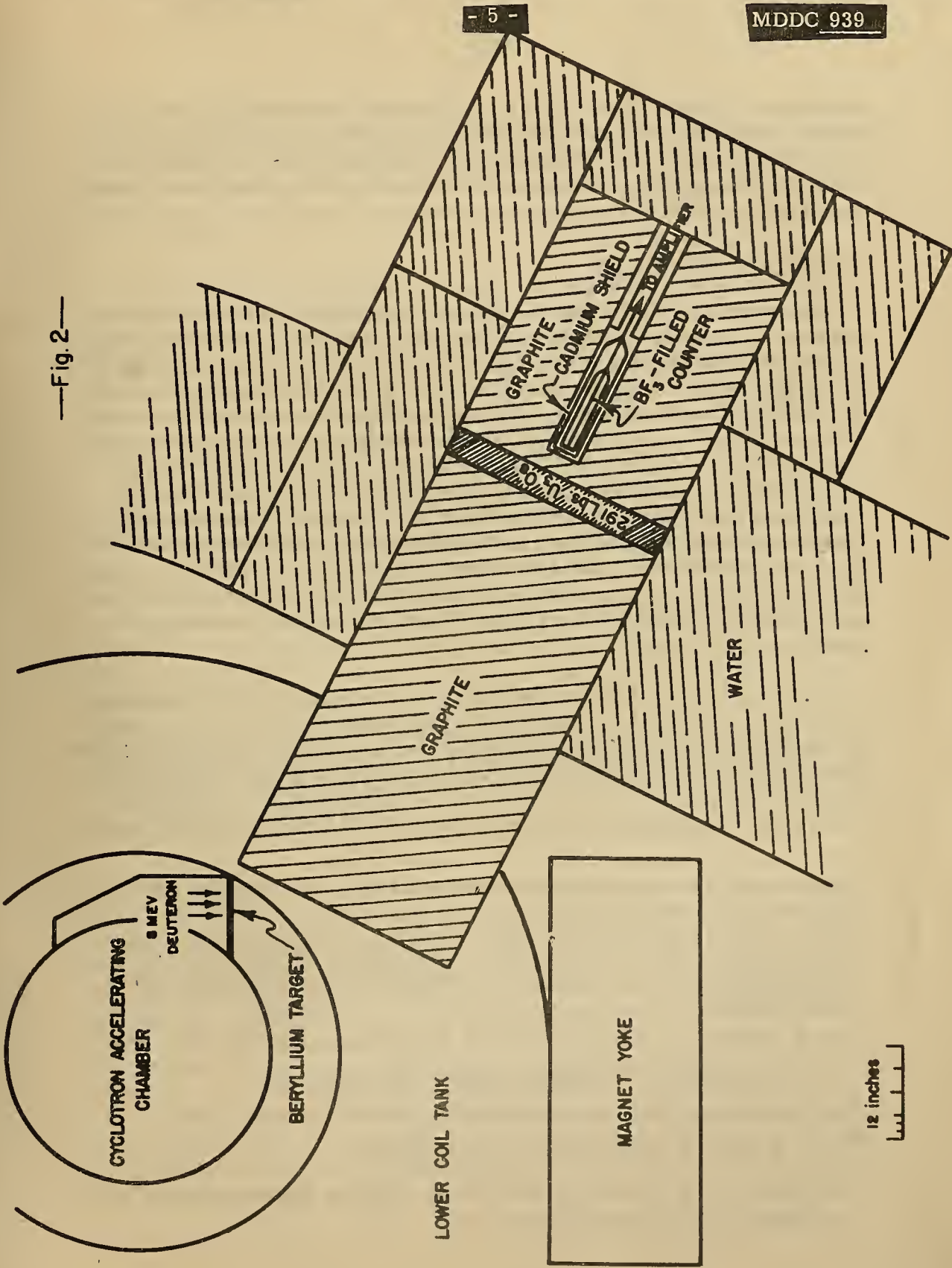
A summary of all of this work gives the following results for the delayed neutron activities for fission of uranium 235:

| Half-life | Relative saturation intensity |
|-----------|-------------------------------|
| 0.4 sec | 0.4 |
| 1.8 " | 0.5 |
| 4.4 " | 1.1 |
| 23 " | 1.0 |
| 56 " | 0.14 |

No activities of longer half-life were observed. In looking for them, activations up to 30 minutes duration were given and the decay of the delayed neutrons was followed for 15 minutes before their activity became indistinguishable from background.

It should be observed that the relative intensities of the various activities as given above may be slightly in error inasmuch as the energy

—Fig. 2—



dependence of their distribution in the paraffin surrounding the counters was not taken into account. The fact that the different activities have different energies has been established.^{6,7} Hughes, Dabbs and Cahn⁶ have since re-surveyed the delayed neutron activities, taking the energy dependence into account.

The Intensity Experiment

The experimental arrangement is indicated in Fig. 2 and may be described as follows: Near the target of the cyclotron a movable graphite column was built, 26" x 30" in cross section, and 96" long. This column was supported in an aperture built into one of the water tanks with which the cyclotron was surrounded. It rode on roller chains so that for use it could be rolled toward the target, between the magnet coils of the cyclotron, until one end was a few inches from the target. Farther back it was completely surrounded by water or other effective shielding from fast neutrons. Five feet from the target end of this column a diaphragm of U_3O_8 was placed; it filled the whole cross section of the column, was 3" thick, and consisted of a sheet iron box containing 291 lbs of U_3O_8 . Along the axis of the column, on the side of the diaphragm away from the target, a large BF_3 counter was placed. It was completely shielded with cadmium 0.020" thick.

The principle of the experiment is now apparent. When the cyclotron was on, the neutrons from the target which penetrated the 5 ft of graphite to reach the U_3O_8 and the counter would be almost all of thermal energies. They, therefore, would cause fissions in the U_3O_8 , but for the most part would not penetrate the cadmium surrounding the counter. On the other hand, many of the neutrons originating in the U_3O_8 would still have enough

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- ¹ H. D. Smyth, Atomic Energy for Military Purposes, Appendix III. Princeton University Press, 1945.
 - ² R. B. Roberts, R. C. Meyer, and P. Wang, Phys. Rev. 55, 510, 1939.
R. B. Roberts, R. C. Meyer, L. R. Hafstad and P. Wang, Phys. Rev. 55, 664, 1939.
 - ³ E. T. Booth, J. R. Dunning and F. G. Slack, Phys. Rev. 55, 876, 1939.
 - ⁴ D. F. Gibbs and G. P. Thomson, Nature 144, 202, 1939.
 - ⁵ K. J. Bostrom, J. Koch and T. Lauritsen, Nature 144, 830, 1939.
 - ⁶ D. J. Hughes, J. Dabbs and A. Cahn, CP-3094.
 - ⁷ M. Burgy, L. A. Pardue, H. B. Willard and E. O. Wollan, MDDC 16, and Phys. Rev. 70, 104, 1946.

energy when reaching the counter nearby to escape capture in the cadmium and be counted. The experiment would, therefore, be to count the "instantaneous" fission neutrons emitted from the U_3O_8 diaphragm with the cyclotron on, then turn off the cyclotron and immediately count the delayed neutrons.

The delayed neutrons were so weak that only a few were registered even after counting instantaneous neutrons at the maximum trustworthy rate permitted by the apparatus. Since the shape of the decay curve was known with moderate precision, it was expedient to measure the intensity of the delayed neutrons by taking the integral number arriving during a certain time interval after turning off the cyclotron, rather than to attempt rate-of-counting measurements.

The start of counting was accomplished through a relay in the input of the scaler which closed when the radiofrequency power contactor of the cyclotron opened. This ensured a uniform, short time interval between the stop of bombardment and the start of counting. The length of this time interval was estimated to be about a fiftieth of a second. A switch was provided which removed the relay from the circuit, thereby making it possible to count when the cyclotron was on. One operator concentrated upon keeping the cyclotron beam steady, and after three minutes of bombardment the other operator went through the following procedure:

- (a) Counted for one minute with the cyclotron on and noted the reading
- (b) Switched the relay into the circuit, and reset the scaler to zero
- (c) Turned off the cyclotron (thereby automatically starting counting) and started a stopwatch
- (d) Counted for 2.0 minutes and noted the number of delayed counts

The bombardment and counting were then repeated. The beam was kept at about $16 \mu A$, which gave a counting rate with the cyclotron on of 15,000 to 20,000 counts per minute. A day's work yielded nineteen satisfactory runs, the main trouble being variability of the cyclotron beam. The delayed neutron counts of these nineteen runs were corrected for the spontaneous fission neutron background (5.0 per min), normalized to a counting rate with the cyclotron on of 10,000 per minute, and then averaged. The mean result was that if, during bombardment, neutrons were arriving at a rate of 10,000 per minute, then in the interval 0.02 sec to 2.0 min after removing the source of primary neutrons, 22.4 ± 3.2 delayed neutrons were counted, where the 3.2 was the probable error of the mean of the nineteen observations. Now the result of the analysis of the decay curves can be expressed as

$$I(t) = C(0.4e^{-1.6t} + 0.5e^{-0.38t} + 1.1e^{-0.16t} + 1.0e^{-0.031t} + 0.14e^{-0.012t}) \quad (1)$$

so that at zero time $I(0) = 3.14C$. The integral count of 22.4 from $t = 0.02$ to $t = 120$ seconds gives from (1) the value $C = 0.46$. Thus $I(0)$ is 1.44 counts per second or 0.86 percent of the counting rate with the cyclotron on.

This result must be modified by two corrections. The more important of these is an upward correction in the delayed neutron intensity, arising from a blank experiment (i.e., with the U_3O_8 removed) in which it was found 365 counts per min per μA were obtained under the same conditions as gave 1180 counts per min per μA with the U_3O_8 present. The resulting factor 1.40 is clearly a considerable overcorrection because it neglects the effect of the absorption of the cyclotron neutrons in the thick layer of U_3O_8 . The other correction arises from missed counts, because of the high counting rates needed to give a reasonable number of delayed counts. It is a downward correction in the relative intensity of the delayed neutrons. The net effect of the two corrections may be estimated at an increase of about 15 percent, leading to the result that in a body of uranium emitting fission neutrons, at equilibrium

$$(1.0 \pm 0.2)\%$$

of the neutrons are delayed by more than 0.02 seconds and have half-lives of 0.4 sec or more. The error indicated here is a guess at the maximum uncertainty arising from the probable error of the counts and the vagueness in the corrections applied. No correction was attempted for a difference in the spatial distribution of the slowed-down fission and delayed neutrons arising from their possibly different initial energies, because of lack of knowledge of the energy spectra. The longitudinal arrangement of the counter would tend to reduce such a correction.

FIGURE CAPTIONS

- Fig. 1. Experimental arrangement used in initial measurement of delayed neutron decay by Snell, Nedzel and Ibser.
- Fig. 2. Diagram of the apparatus used to measure the intensity of the delayed neutrons relative to that of the instantaneous fission neutrons.

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